

CLAIMS

We claim:

- 1 1. (Currently amended) A polymer electrolyte comprising:
2 a modified ~~polymeric material, said modified polymeric material including a~~
3 halogen containing polymer having an enhanced halogen level, ~~said enhanced halogen level~~
4 relative to a halogen content of an unmodified ~~said~~ halogen containing polymer formed from
5 polymerization of its monomer;
6 a salt of an alkali metal; and
7 an aprotic solvent, wherein said salt and said aprotic solvent are integrated with
8 said modified polymeric material as a homogeneous material.

- 1 2. (Currently amended) The polymer electrolyte of claim 1, wherein said unmodified
2 halogen containing polymer includes at least one chlorine containing polymer.

- 1 3. The polymer electrolyte of claim 2, wherein said chlorine containing polymer is
2 polyvinylchloride (PVC).

- 1 4. (Currently amended) The polymer electrolyte of claim ~~[[3]]~~ 1, wherein said
2 polyvinylchloride (PVC) is ~~suspension polyvinylchloride (PVC)~~ a lithium ion conductivity of
3 said polymer electrolyte at 25 C is between 0.01 S/cm² and .108 S/cm².

1 5. (Currently amended) The polymer electrolyte of claim [[3]] 1, wherein said
2 ~~polyvinylchloride (PVC) is emulsion polyvinylchloride (PVC)~~ a lithium ion conductivity of said
3 polymer electrolyte at 25 C is between 0.066 S/cm² and .108 S/cm².

1 6. The polymer electrolyte of claim 1, wherein said modified polymeric material
2 comprises C-PVC, said C-PVC having 60-72 wt % chlorine.

1 7. The polymer electrolyte of claim 6, wherein said polymer electrolyte comprises
2 10-40 wt % of said C-PVC.

1 8. The polymer electrolyte of claim 1, wherein said alkali metal salt is at least one
2 selected from the group consisting of LiClO₄, LiBF₄, LiAsF₆, LiPF₆, LiCF₃SO₃ and
3 LiN(CF₃SO₂)₂.

1 9. The polymer electrolyte of claim 1, wherein said electrolyte comprises from 3-20
2 wt % of said salt of an alkali metal.

1 10. The polymer electrolyte of claim 1, wherein as said aprotic solvent is at least one
2 selected from the group consisting of propylene carbonate, ethylene carbonate, dimethyl
3 carbonate, gamma-butyrolactone, 1,3-dioxolane and dimethoxyethane.

{WP171485;1}

1 11. The polymer electrolyte of claim 1, wherein said electrolyte comprises 40-82 wt
2 % of said aprotic solvent.

1 12. (Currently amended) A rechargeable battery, comprising:
2 an anode containing an alkali metal;
3 a cathode; and
4 a polymer electrolyte formed from a modified ~~polymeric material, said modified~~
5 ~~polymeric material including~~ a halogen containing polymer having an enhanced halogen level;
6 ~~said enhanced halogen level~~ relative to a halogen content of an unmodified said halogen
7 containing polymer formed from polymerization of its monomer, a salt of an alkali metal and an
8 aprotic solvent wherein said salt and said aprotic solvent are integrated with said modified
9 polymeric material as a homogeneous material.

1 13. (Currently amended) The rechargeable battery of claim 12, wherein said
2 unmodified halogen containing polymer comprises at least one chlorine containing polymer.

1 14. The rechargeable battery of claim 13, wherein said modified polymeric material
2 comprises chlorinated polyvinylchloride (C-PVC).

1 15. The rechargeable battery of claim 12, wherein in said anode comprises lithium.

1 16. (Currently amended) The rechargeable battery of claim 12, wherein said anode
2 ~~comprises a lithium alloy~~ a lithium ion conductivity of said polymer electrolyte at 25 C is
3 between 0.01 S/cm² and .108 S/cm².

1 17. (Currently amended) The rechargeable battery of claim 16, wherein a lithium ion
2 conductivity of said polymer electrolyte at 25 C is between 0.066 S/cm² and .108 S/cm² ~~as said~~
3 ~~lithium alloy is at least one selected from the group consisting of lithium-aluminum, lithium-~~
4 ~~aluminum-silicon, lithium-aluminum-cadmium, lithium-aluminum-bismuth and lithium-~~
5 ~~aluminum-tin.~~

1 18. (Currently amended) The rechargeable battery of claim 12, wherein said anode
2 comprises a lithium-ion intercalation material.

1 19. The rechargeable battery of claim 12, wherein said cathode comprises a metal
2 oxide.

1 20. The rechargeable battery of claim 12, wherein said cathode comprises a lithium-
2 transition metal oxide.

1 21. The rechargeable cell of claim 12, wherein said cathode is at least one selected
2 from the group consisting of MnO₂, LiMn₂O₄ and vanadium oxides (V_xO_y).

1 22. The rechargeable cell of claim 12, wherein said cathode comprises a organic
2 polymer.

1 23. The rechargeable cell of claim 12, wherein said cathode is at least one selected
2 from the group consisting of polyviologen, polyacetylene and polypyrrole.

1 24. The rechargeable cell of claim 12, wherein said cathode comprises a sulfur
2 containing material.

1 25. The rechargeable cell of claim 12, wherein said cathode is at least one selected
2 from the group consisting of TiS_2 , S, polysulphide and polythiophene.

1 26. (Cancelled)

1 27. (Cancelled)

1 28. (Cancelled)

1 29. (Cancelled)

1 30. (Cancelled)

1 31. (Cancelled)

1 32. (Cancelled)

1 33. (Cancelled)

1 34. (Cancelled)

35. (Cancelled)

36. (Cancelled)

Reply

This Reply is responsive to an Office Action dated February 12, 2004 and is filed along with a petition for a one-month extension of time and authorization to charge the required statutory fee for extension.

There were 36 claims filed with application. Pursuant to a restriction requirement, Applicants elected to prosecute group 1, claims 1-26, with traverse, during a telephone conference with the Examiner on February 4, 2004. Applicants herein affirm their election to prosecute group 1, claims 1-26.

In this Reply, claims 1, 2, 4, 5, 12, 13, and 16-18 have been amended and claims 26-36 have been cancelled. No new matter has been added.

Claims 1-26 were rejected under 35 U.S.C. §112, first paragraph, based on the Examiner's assertion that the specification, while being enabling for a chlorine containing polymer having an enhanced chlorine level, does not reasonably provide enablement for any halogen containing polymer having an enhanced halogen level. Applicants respectfully disagree with the 35 U.S.C. §112, first paragraph rejections for reasons described below.

Applicants initially note that it is well known that the chemical properties of the various halogens (e.g. F, Cl, Br, I) are quite similar, and thus generally interchangeable. The vinyl chloride monomer is commonly polymerized to form PVC. The vinyl bromide monomer is well known and is commonly used to form polyvinyl bromide (PVB) chloride. The vinyl fluoride monomer is also well known and is used to form polyvinyl fluoride (PVF). PVB and PVF have properties similar to PVC, such as solvent solubility. Accordingly, even though Applicants' specification does not provide examples of specific

polymeric materials containing halogens other than chlorine, one having ordinary skill in the art in possession of Applicants specification would recognize the invention can be practiced with halogen containing polymers where the halogen is other than chlorine, and be able to practice the invention using these polymers without undue experimentation.

Moreover, Applicants' application teaches use of halogenation (increasing halogen content in the structure of polymer material) to increase the amorphization of polymer matrix structure (see for example, page 14, lines 11 and 12; page 15 line 22 to page 16 line 1). Amorphous structure of polymer materials, as it is known in the art, is more preferable for charge transfer (promotes an ionic conductivity increase). Although PVC is used as an exemplary polymer to describe the invention, one having ordinary skill in the art would recognize that the invention applies to clearly applies to other crystallized halogenated polymers, such as polyvinylidene fluoride (PVF). Crystallinity level of such polymers can be up to 40% (for PVC ~10%), that being one of the disadvantages of these polymer matrixes. Additional fluorination in the case of polyvinylidene fluoride to render the resulting material more amorphous and as a result better for forming polymer electrolytes according to the teachings of the invention would be available to a person having ordinary skill in the art, such as by the processes below which are identical to the heterogeneous or homogeneous processes for forming C-PVC disclosed on page 21, lines 3-10, except where F_2 has replaced Cl_2 and PVF is substituted for PVC:

Under a heterogeneous fluorination process, for example, gaseous fluorine (F_2) would be passed through a suspension of powdered polyvinylidene fluoride (PVF) in a liquid medium, such as water or CCl_4 . Under homogeneous fluorination, gaseous

fluorine would be passed through polyvinylidene fluoride (PVF) in a solution of one or more organic solvents.

In light of the above, Applicants submit it is clear that the 35 U.S.C. §112, first paragraph rejection of claims 1-26 should be removed.

The Examiner also rejected claims 1-26 under 35 U.S.C. §112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. Claims 1, 2, and 13 have been amended consistent with the suggestion from the Examiner to now refer to a "modified halogen containing polymer" and an "unmodified halogen containing polymer", while claim 18 now adds the word intercalation as suggested by the Examiner. Accordingly, Applicants submit that the 35 U.S.C. §112, second paragraph rejection of claims 1-26 is now overcome.

Before reviewing the claim rejections based on art, Applicants will first review the claimed invention as now recited in amended claim 1. Amended claim 1 recites a polymer electrolyte comprising a modified halogen containing polymer having an enhanced halogen level relative to a halogen content of an unmodified halogen containing polymer formed from polymerization of its monomer. The electrolyte includes a salt of an alkali metal and an aprotic solvent. *The salt and aprotic solvent are integrated with the modified polymeric material as a homogeneous (single phase) material.*

The homogeneous (single phase) polymeric electrolyte structure results from Applicants' novel process for forming the electrolyte where the electrolyte is dissolved in solution together with the Li salt and two solvents (forming a solution including all electrolyte components), such as disclosed on page 21, lines 11-15 (copied below):

The C-PVC formed can then be combined with LiClO_4 and propylene carbonate which are together dissolved in tetrahydrofuran (THF) to form a *substantially homogenous solution*. This solution is then casted upon a glass sheet or placed directly on the electrode and dried 24 hours at room temperature and then for 48 hours under a vacuum at 45°C . After drying the thin C-PVC SPE film, the film is ready for use in lithium batteries.

Since the polymer, the salt, and the solvent are homogeneously mixed in solution, following evaporation of the volatile solvent (THF), the resulting electrolyte material is homogeneous. The homogeneous single phase nature of Applicants' claimed electrolyte which comprises the polymer, the salt and some aprotic solvent being intimately mixed is also evidenced by the high electrical conductivity values obtained by Applicants, such as the lithium ion conductivity of up to $.108 \text{ S/cm}^2$ disclosed in the application (Example 6, page 26, line 2). For a $100 \mu\text{m}$ thick electrolyte layer, the resulting Li ion conductivity would be $1.08 \times 10^{-3} \text{ S/cm}$. The lithium ionic conductivity obtained is much higher than the Li ion conductivity provided by conventional polymer electrolytes (such as Chang and Chia cited by the Examiner) which are non-homogeneous two-phase materials which have the pores of the polymer stuffed with an electrolyte/salt solution.

Now turning to rejections based on art, claim 1 was rejected under 35 U.S.C. §102(b) as being anticipated by U.S. Patent No. 5,389,463 to Chang et al. ("Chang"). According to the Examiner, "Chang '463 teaches a polyvinyl halide having a halogen content of at least about 55 wt% of the polymer."

Chang is entitled "Battery Separator". Chang's separator includes a microporous sheet product having first and second major surfaces and a thickness of less than about 50 mils, formed from a uniform mixture of a halogenated polyolefin polymer and a filler or a halogenated polyolefin polymer and surfactant/filler agent. The sheet product has a

fibrous sheet embedded within the mixture and between the first and second major surfaces. Chlorinated PVC is one of the polymers disclosed.

Chang only discloses a porous separator. The porous separator does not have any significant ionic conductivity. To produce an electrolyte for use in a battery, the porous separator must be impregnated with a liquid electrolyte, where the electrolyte becomes disposed in the pores of the separator. The electrolyte would then provide Li ion conductivity. As a result, Chang's hypothetical impregnated separator would consist of two separate phases analogous to Chia (discussed below):

- i. The first phase being a solid porous separator
- ii. The second phase being a liquid electrolyte.

Such a two-phase system is clearly distinct as compared to Applicants' claimed homogeneous electrolyte including the salt, electrolyte and polymer intermixed as a single phase material. As a result, the two-phase system has a number of significant disadvantages. First, the electrolyte will have comparatively low ionic conductivity, since ionic conductivity is realized only by the liquid phase which is disposed in the pores of the separator. Second, insufficient contact with the solid electrode surface and the possibility of loss the contact during power source work is expected. As the result of the second disadvantage, it is generally necessary to add additional free volume of electrolyte to help ensure contact with the surface of solid electrode.

In contrast to Chang, the claimed invention recited in amended claim 1 is a one-phase homogeneous electrolyte system. The electrolyte claimed is not a mechanical mixture or component composition. The electrolyte according to the claimed invention is new chemical structured material that is demonstrated by the high ionic conductivity

values disclosed in Applicants' application, such as claimed in amended claims 4 and 5 of between 0.01 S/cm² and .108 S/cm² and between 0.066 S/cm² and .108 S/cm², respectively.

Accordingly, Applicants submit that amended claim 1 and its respective dependent claims are patentable over Chang. The rechargeable battery recited in amended claim 12 includes the patentable electrolyte recited in amended claim 1.

Accordingly, Applicants submit that amended claim 12 and its respective dependent claims are patentable over Chang.

Claims 1-6, 8, 10, 12-15, 18-22, were determined to be anticipated by U.S. Patent No. 6,617,078 to Chia et al., ("Chia"). According to the Examiner, "Chia teaches a lithium ion rechargeable battery having a negative electrode, a positive electrode and a separator/polymer electrode there between comprising a chlorinated polymer."

Chia discloses production of the electrolyte in two steps. A micro porous film separator is formed by mixing of chlorinated PVC and terpolymer of vinylidene chloride. The terpolymer of vinylidene chloride is used to increase the physical mechanical properties of separator, since the film based on C-PVC is disclosed to be brittle. According to the fourth paragraph of the detailed description:

Preferably, the chlorinated PVC is blended with a terpolymer of vinylidene chloride. It showed enhanced high temperature stability and also displayed mechanical integrity in the as-cast and extracted separator films. If using vinylidene chloride terpolymer alone as the polymer binder, the as-cast separator shows good mechanical properties, but it becomes very brittle with poor handability after the removal of plasticizer, a step used to produce porous membrane. Separately, if using chlorinated PVC alone as the polymer binder, the as-cast separator film appears to be tacky and hence limits its application. It is the blending of chlorinated PVC and terpolymer of vinylidene chloride, which provides the most desirable mechanical properties.

The separator is impregnated with a 1M solution of LiPF_6 in ethylene carbonate and dimethyl carbonate-based electrolytes. A two phase electrolyte is clearly formed, where the electrolytes fill the porosity of the polymer. No Li ion conductivities are disclosed by Chia.

In contrast to Chia's two-phase electrolyte, Applicants' claimed electrolyte is a homogeneous single phase electrolyte where all the components are blended together resulting from Applicants novel process described above where all electrolyte components are in solution together. Due to Chia's two-phase electrolyte structure, Chia cannot obtain Applicants' high Li ion conductivity values claimed in amended claims 4 and 5 of between 0.01 S/cm^2 and $.108 \text{ S/cm}^2$ and between 0.066 S/cm^2 and $.108 \text{ S/cm}^2$. Accordingly, Applicants submit that amended claim 1 and its respective dependent claims are patentable over Chia.

As noted above, the rechargeable battery recited in amended claim 12 includes the patentable electrolyte recited in amended claim 1. Accordingly, Applicants submit that amended claim 12 and its respective dependent claims are patentable over Chia.

Claims 1-26 were rejected under 35 U.S.C. 103(a) as being unpatentable over U.S. Patent No. 5,252,413 to Alamgir et al. ("Alamgir") in view of Chang and/or in view of Chia. According to the Examiner, "Alamgir teaches a lithium battery using lithium ion conductive solid polymer electrolytes composed of solvates of lithium salts immobilized in a solid organic polymer matrix." The Examiner asserts that:

at the time the invention was made one of ordinary skill in the art would have been motivated to substitute the PVC of Chia or Chang for the PVC of Alamgir in order to improve the properties of the battery. (... battery and mechanical integrity of the separator/polymer (Chia); Chang teaches chlorinated PVC has high tensile strength and ductility).

Applicants respectfully disagree with the motivation to combine noted above. Alamgir discloses lithium batteries using Li ion (Li⁺) conductive solid polymer electrolytes derived by immobilizing solvates formed between a Li salt and an aprotic organic solvent (or mixture of such solvents) in PVC. The PVC used is conventional PVC, not the modified halogen containing polymer claimed by Applicants' having an enhanced halogen level claimed by Applicants.

Alamgir's invention is directed solely at the formation of high Li ion conductivity electrolytes and resulting batteries thereof for use at room temperature (not requiring heating for sufficient Li ion conductivity). The 25 C Li ion conductivities achieved are quite high, such as at greater than $10^{-3} \text{ ohm}^{-1} \text{ cm}^{-1}$ at room temperature (see second paragraph; detailed description). These impressive results which approach the extremely high ion conductivities demonstrated by Applicants, are achieved using unmodified PVC.

Accordingly, one having ordinary skill in the art would not look to C-PVC disclosed in Chang or Chia in an attempt to improve mechanical properties of Alamgir's electrolyte. First as noted above, Chia teaches away from use of C-PVC because it is taught as being brittle and tacky and thus requiring a co-polymer, such as the terpolymer of vinylidene chloride. Although it may improve mechanical integrity (reduce brittleness), addition of the terpolymer of vinylidene chloride would likely reduce ionic conductivity and complicate the process to form the electrolyte. Moreover, Alamgir reports successful cycling results with no mention of mechanical problems. Finally, the lack of reported Li ion conductivities reported by Chia suggests marginal results while the expected low Li ion conductivity obtained from a hypothetical electrolyte based on Chang which would indicate to one having ordinary skill in the art at the time of the

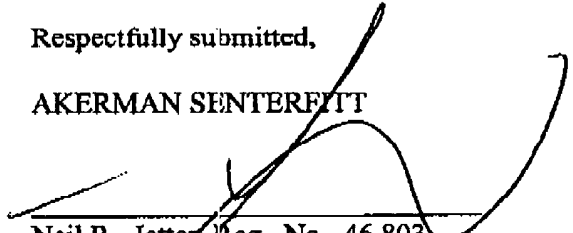
invention that use of C-PVC instead of conventional PVC would degrade the $10^{-3} \text{ ohm}^{-1} \text{ cm}^{-1}$ at room temperature Li ion conductivities provided by the electrolytes disclosed by Alamgir. Accordingly, Applicants submit that amended claim 1, amended claim 12, and their respective dependent claims are patentable over Alamgir in view of Chang and/or in view of Chia.

Applicants have made every effort to present claims which distinguish over the prior art, and it is believed that all claims are in condition for allowance. However, Applicants invite the Examiner to call the undersigned (direct 561-671-3662) if it is believed that a telephonic interview would expedite the prosecution of the application to an allowance.

Respectfully submitted,

AKERMAN SENTERFITT

Date: June 14, 2004



Neil R. Jetter, Reg. No. 46,803
222 Lakeview Avenue, Suite 400
P. O. Box 3188
West Palm Beach, FL 33402-3188
(561) 653-5000